

REMARKS

Claims 1-9 currently are pending. Claim 1 currently has been amended.

The present invention relates to processes for the preparation of catalysts having low volume shrinkage. Further, the present invention relates to processes for the steam reforming of methanol and of higher alcohols, for the partial oxidation of hydrocarbons for hydrogen productions and CO shift reactions, in which these catalysts are used.

The catalysts according to the present invention contain passivated copper and zinc oxides and/or alumina and are prepared by : (1) precipitating a mixture of catalyst precursor components dissolved or suspended in a diluent with anion-containing precipitating agents, washing and drying to form a solid catalyst precursor in the form of powder or granules, (2) calcining the solid catalyst precursor obtained in stage (1) to an anion content from the precipitating agent of from 0.1 to 2.5% by weight by choice of the temperature and the duration of the calcination and, (3) shaping and, if required, reducing and passivating the calcine catalyst precursor from stage (2) in any desired order to form the catalyst.

The catalysts prepared by the novel process have a volume shrinkage during or after operation of less than 5% in combination with high mechanical strength during operation; see page 4, lines 28-31 of the present specification.

Both are achieved by calcining a catalyst precursor comprising basic metal salts in such a way that a defined amount of residual anions remains in the catalyst. This can be controlled by choice of temperature and duration for the calcinations which is

now clarified in the new set of claims.

Claims 1-9 are rejected under 35 USC § 103(a) as being unpatentable over Williams (US 5,928,985) in view of Kumberger et al. (US 6,051,163).

Williams relates to copper catalysts which are stabilized by a special passivating technique. The catalysts, which are used according to Williams are prepared by dry mixing the copper compound with any other desired ingredients of the precursor (Zn, Cr, Zr, Mg, Si and/or Al) or by precipitating from a copper salt solution with a suitable basic compound.

Generally, after forming such a composition containing the copper compound and other ingredients, the precursor is calcined to convert the copper compound. Detailed information about this calcinating step is not disclosed in Williams. In particular, examples 1 and 3 of Williams are silent as to the calcinating step in general, because they only describe the passivating procedure starting from pelleted copper catalyst precursors which are already calcined.

Calcination steps in the process for the preparation of catalysts are usual and known to persons of ordinary skill in the art. During a calcination step metals are converted into their oxides as a result of heating to a high temperature. Thereby, a person of ordinary skill in the art only considers reaching a temperature which is high enough for the aforementioned conversation by avoiding a temperature which is too high so that the catalyst may be irreversibly damaged. Other considerations are not kept in mind. Especially, the duration of calcinations and the exact control of the

temperature within the aforementioned limits are of small importance.

In contrast to this, the present invention teaches to carry out the calcinations at a special temperature and during a special duration so that the solid catalyst precursor is calcined to an anion content from the precipitated agent of from 0.1 to 2.5% by weight. Thereby, it is not possible to define the calcination temperature and the duration of the calcination in detail because the parameters may change depending on the catalyst system.

Thus, catalysts precipitated by means of alkaline generally require higher temperatures to achieve the desired effect. The temperatures also depend on the furnace used (muffle furnace in the laboratory, rotary tubular furnace in industry) and may often be the temperatures set on the controller, it being possible for the internal temperatures of the furnaces, in particular in the case of rotary tubular furnaces, sometimes to deviate substantially therefrom. In addition to the temperature of the calcination, the duration of the calcination may also be important for the performance of the prepared catalyst. Here however, the window is dependent on the throughput of the industry rotary tubes and is preferably 0.1 to 1, in particular from 0.2 to 0.6 hour, generally in the region of one hour.

This is shown in the examples of the present invention: Example 1 (page 9 of the present specification) covers a copper catalyst which is prepared analogously to EPA0296734. Thereby, it is shown that a calcination has to be done by a temperature of from 500 °C for one hour to reach at an anion content of 0.3% by weight (catalyst 1c)

while calcinations at temperatures of from 300 to 400 °C are not sufficient enough to reach the proposed anion content (catalysts 1a and 1b).

In example 2 of the present application (page 10 of the present specification) it is shown that a copper catalyst which is prepared according to DEA3520832 has to be calcined at a temperature of from 430 °C to produce a catalyst with an anion content of from 0.3% by weight. Calcinations at temperatures of from 300 to 375 °C are not sufficient enough.

These examples show that the calcination temperatures and the duration of the calcinations are critical features to arrive at catalysts with an anion content of less than 2.5% by weight. The exact parameters have to be adapted to the catalytic system, see above.

Because there is no temperature and no duration of the calcination step indicated in Williams, it would not have been obvious to one of ordinary skill in the art to have obtained an anion content in the instantly claimed range only on the basis of the disclosure of the prior art. In fact, the skilled person is taught to carry out the calcination in a way so that the metals of the catalyst precursor are converted into their oxides. According to prior art, the exact parameters for this conversation are not critical and can be chosen in a broad range. In contrast, the present application shows that the calcination parameters are critical and have to be maintained exactly.

Also, the subject matter of the present invention is also not obvious by a combination of Williams and Kumberger et al. This is because Kumberger also fails to

show the feature that the solid catalyst precursor obtained in stage (1) is calcined to an anion content from the precipitated agent of from 0.1 to 2.5% by weight by choice of the temperature and the duration for the calcination.

The object of Kumberger et al. is to provide catalysts for steam reforming methanol which have a high catalyst activity, based on the catalyst volume, and are thus usable with advantage, in particular, for mobile applications in fuel cells.

The calcination, which is achieved in the process according to Kumberger et al. comprises a calcinating step. This calcinating step is explained in detail in column 3, lines 15-18 of Kumberger et al. There, it is mentioned that the calcination of the precipitate is done at a temperature from 250-800 °C to form a mixed oxide. Nowhere in Kumberger et al. is it mentioned that the calcination of the catalysts is done to an anion content from the precipitating agent of from 0.1-2.5% by weight.

Also, the mentioned temperature range in Kumberger et al. is too broad and too extensive to suggest that the calcination step is done to an anion content from the precipitated agent of from 0.1 to 2.5% by weight by choice of the temperature and the duration for the calcination. As can be seen from the examples of the present invention, the calcination temperature and the calcination duration are critical features and have to be maintained in a small range which depends on the catalytic system. In contrast to this, Kumberger et al. teaches to use conditions in a temperature range of 250 to 800 °C and without any information about the duration. Therefore, Kumberger et al. also fails to have an exact technical teaching which leads to the inventive solution.

The non-obviousness of the subject matter of the present invention is also highlighted by the fact that the question is not whether the skilled person in the art could have carried out the invention and could have reached the proposed anion content but rather whether he would have done so in the hope of solving the underlying technical problem or in the expectation of some improvement or advantage. So the point is not whether the skilled person could have arrived at the invention by modifying the prior art (e.g. by choosing a defined temperature and a defined calcination duration), but rather whether- in expectation of the advantages actually achieved (i.e. in the light of the technical problem addressed)- he would have done so because of the motivation to combined in the prior art.

In both references, there is no motivation to combine the references as the examiner has done. The references deal with different technical problems and nowhere is it stated that calcining the solid catalyst precursor obtained in stage (1) to a special anion content from the precipitated agent leads to catalysts having a volume shrinkage during or after operation of less than 5% in combination with high mechanical strength during operation. Moreover, the special anion content is not mentioned literally Williams or Kumberger et al. Therefore, a person of ordinary skill in the art does not get any information or a pointer into the direction of the present invention.

Claims 1-9 are rejected under 35 USC § 103(a) as obvious over copending 10/090,762.

10/090,762 relates to processes for the preparation of catalysts having low

volume shrinkage and high mechanical hardness. Furthermore, 10/090,762 relates to processes for the steam reforming of methanol and of higher alcohols, for the partial oxidation of hydrocarbons for hydrogen productions and CO shift reactions, in which these catalysts are used.

The catalysts according to 10/090,762 contain passivated copper and zinc oxides and/or alumina and are prepared by (1) precipitating or spray-drying a mixture of catalyst precursor components dissolved or suspended in a diluent in order to form a solid catalyst precursor in the form of powder or granules, (2) calcining and reducing the solid catalyst precursor obtained in stage (1), (3) passivating the reduced catalyst precursor obtained in stage (2), and (4) shaping the passivated catalyst precursor obtained in stage (3) to form the catalyst.

The catalysts prepared by Williams have low volume shrinkage during or after operation and high mechanical stress. Both are achieved by reducing a catalyst precursor with hydrogen during the preparation itself and then passivating it again with oxygen in the specific proposed manner.

Therefore, Williams concerns the same technical problem but uses a different solution compared to the present application. While 10/090,762 teaches to reduce the catalyst precursor with hydrogen during the preparation itself and then passivating it again with oxygen in the specific proposed manner, the present application teaches to calcine the solid catalyst precursor obtained in stage (1) to an anion content from the precipitated agent of from 0.1 to 2.5% by weight by choice of temperature and the

duration of the calcinations.

Also, 10/090,762 mentions the calcinations procedures with broad parameters (e.g., by two calcination temperatures above and lower than 300 °C) without any promptings into the direction of the present inventive solution.

Claims 1-7 and 9 are provisionally rejected under the judicially created doctrine of obviousness-type double patenting as being unpatentable over claims 1-8 of copending application 10/090,762.

The determination of obviousness-type double patenting essentially involves a determination of non-obviousness under 35 USC § 1032, with the exception that the patent disclosure is not applicable as prior art and corresponding to a determination of obviousness under 35 USC § 103, the invention as a whole must be considered when an analysis of obviousness-type double patenting is made.

The invention disclosed and claimed in 10/090,762 relates to a catalyst which is obtained by (1) precipitating or spray drying a mixture of catalyst precursor components dissolved or suspended in a diluent to form a solid catalyst precursor in the form of powder or granules, (2) calcining and reducing the solid catalyst precursor obtained in stage (1), (3) passivating the reduced catalyst precursor obtained in stage (2) and (4) shaping the passivated catalyst precursor obtained in stage to form the catalyst.

Accordingly, a precursor powder or granulate is first calcined, the calcined intermediate is reduced and subsequently the reduced intermediate is passivated. Only after the precursor powder or granulate has been pre-treated in the requisite manner

(stages (1) to (3)) the catalyst is formed. More particularly, it was found that a catalyst which is prepared in the specified manner exhibits particular properties which are not found in a catalyst obtained by calcining a precursor powder or granulate and subsequently shaping the calcined precursor. The distinct and significant impact on the catalyst properties which results from the particular manner in which this catalyst is obtained is illustrated by the experiments summarized on pages 13-16 of the application.

In example 1 which illustrates the invention an initial Cu catalyst precursor was first calcined, then reduced and subsequently passivated before it was shaped to tablets. For comparison purposes, the calcined initial Cu catalyst precursor was shaped to tablets without a prior reduction and passivation stage in comparative example 1. The properties of the catalysts obtained in example 1 and in comparative example 1 were then investigated in a methanol reforming process. The catalyst test showed that the catalyst of example 1 suffered a loss of compressive strength from an initial value of 39.5 N/tablet to 34.2 N/tablet, corresponding to 5.3 N/tablet, whereas the catalyst of comparative example 1 suffered a loss of compressive strength from an initial value of 51.0 N/tablet to 7.4 N/tablet, corresponding to 43.6 N/tablet. The catalyst according to applicants' invention is, accordingly, by far less prone to loose compressive strength than the comparative catalyst. In addition to the distinct improvement of the compressive strength of the catalyst which is achieved in accordance with applicants' (10/090,762) invention, applicants' (10/090,762) catalyst

also has a distinct advantage over the comparative catalyst where the volume shrinkage is concerned. While the catalyst of example 1 suffered a volume shrinkage of 12.5 %, the catalyst of comparative example 1 suffered a volume shrinkage of 22.4%.

For the comparison in comparative example 2, applicants of 10/090,762 repeatedly passivated and reduced the catalyst of comparative example 1 before the comparative catalyst was employed the methanol reforming process. The comparative catalyst employed in comparative example 2 was, accordingly, obtained by first calcining the precursor, then shaping the calcined precursor to tablets and subsequently reducing and passivating the tablets. The comparative catalyst which was obtained in that manner exhibited a compressive strength of only 3.5 N/tablet, as compared to the compressive strength of 39.5 N/tablet exhibited by catalyst of example 1 representing applicants' invention.

When the comparative catalyst and the catalyst according to applicant's invention is illustrated in example were tested for their stability in the methanol reforming process, the compressive strength of the comparative catalyst was further reduced to 2.4 N/tablet, corresponding to a loss of compressive strength of about 32%, whereas the compressive strength of the catalyst of example 1 was reduced to 34.2 N/tablet, corresponding to a loss of compressive strength of about 14%.

In contrast to the above invention which provides for an improvement of the catalyst compressive strength by employing specific procedure steps in a particular

order, the present application 10/090,763 relates to a catalyst wherein the hardness is adapted by means of controlling the anion content of the catalyst. With regard to the provisions in claim 1 the present application "shaping and, if required, reducing and passivating the calcined catalyst precursor from stage (ii)" may be conducted "in any desired order." According to the teaching of the present application (10/090,763), it is therefore not critical at which stage of the catalyst preparation a reduction and subsequent passivation is conducted, and a person of ordinary skill would expect that the point in the preparation of catalyst at which the reduction and passivation is conducted is of no consequence with regard to the catalyst properties. Therefore, 10/090,762, does not render obvious the present claims. Favorable reconsideration of the examiner's position and withdrawal of the provisional obviousness-type double patenting rejection is therefore respectfully solicited.

It is requested that a three month extension of time be granted.

Applicants also herein submit a declaration pursuant to 37 CFR. 1.132.

Please charge any shortage in fees due in connection with the filing of this paper, including Extension of Time fees to Deposit Account No. 11-0345. Please credit any excess fees to such deposit account.

Respectfully submitted,
KEIL & WEINKAUF

A handwritten signature in black ink, appearing to read "Daniel Kim", written over the printed name.

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